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Life cycle assessment comparing the treatment of surplus activated sludge in a sludge treatment reed bed system with mechanical treatment on centrifuge

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ABSTRACT

In Denmark, the conventional method for treating sewage sludge is mechanical dewatering and subsequent storage. However, sludge treatment reed bed systems, which are holistic sludge treatment facilities combining the dewatering, mineralisation and storage of sludge, have been more common during the last three decades. Treatment of sludge in a sludge treatment reed bed system can be combined with post-treatment (further dewatering and mineralisation) on a stockpile area. This study aimed to compare the environmental performances of a mechanical sludge treatment method with the sludge treatment reed bed system strategy, using the life cycle assessment approach and a life cycle inventory based on newly generated data obtained from Danish reference facilities. The scenarios based on the different treatment methods were initiated by sludge entering the sludge treatment reed bed system or the centrifuge and terminated by land application of the final sludge product. The environmental impacts caused by the sludge treatment reed bed system strategy were comparable to or lower than those caused by the mechanical sludge treatment method. The impacts on climate change were the same for all the treatment scenarios; however, the conversion of organic carbon and nitrogen into gas species was more efficient in the sludge treatment reed bed system compared to mechanical treatment. Thus, mechanically treated sludge contained more nitrogen, causing higher nitrogen emissions (nitrous oxide, nitrate and ammonia) when applied on land. Furthermore, the impact of resource depletion was higher for mechanical dewatering due to a larger fossil fuel demand related to daily operation and longer transportation distances in this scenario. According to the results of the life cycle assessment, there were no considerable environmental gains made by combining the treatment of sludge in a sludge treatment reed bed system with post-treatment on a stockpile area. However, some practical aspects, which are not expressed in a life cycle assessment, should also be taken into consideration when evaluating the performances of sludge treatment scenarios.

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1. Introduction

Sludge Treatment Reed Bed (STRB) systems have been used for treating sludge in Denmark since 1988 (Nielsen et al., 2014). The STRB system treatment method is also employed in other European countries, e.g. France (Vincent et al., 2011), Italy (Peruzzi et al., 2013), Spain (Uggetti et al., 2009) and United Kingdom (Nielsen and Cooper, 2011). An STRB system is a holistic sludge treatment facility that combines the dewatering, mineralisation and storage of sludge. These systems are often used for the treatment of sludge originating from domestic wastewater treatment, but they are also used to treat other types of sludge, e.g. from waterworks (Nielsen and Cooper, 2011) or aqua cultural sludge (Summerfelt et al., 1999). Commonly, an STRB system is built as a part of a wastewater treatment plant (WWTP) and receives the sludge produced at this unit. An STRB system consists of a number of beds, often eight, 10 (Nielsen and Willoughby, 2005) or more, to which sludge is applied over several years. While retained in the beds, the sludge is gradually dewatered and mineralised. After 8–12 years of dewatering and mineralisation, the final sludge product (the sludge

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residue left at the end of the entire treatment and storage processes) is excavated and applied to agricultural land as fertiliser and soil improvement.

Since STRB systems were introduced in Denmark, it has been common practice to empty beds during late summer or early autumn and then transport the excavated sludge residue directly to agricultural land after harvest. However, in recent years, a new procedure has been employed by some STRB systems: The beds are emptied in spring and the excavated sludge residue immediately transferred to a stockpile area at the WWTP where it undergoes post-treatment (further dewatering and mineralisation) until autumn. During post-treatment, the sludge residue undergoes further mineralisation and dewatering due to increased evaporation. This approach has the advantage that the emptied beds can be put back into operation in summer, as the reeds will regrow during spring/early summer. If the excavation happens in autumn, the emptied bed must still rest until next coming spring/summer, as the reeds remain dormant during autumn and winter. Originally, stockpile areas were of a simple design, namely an outdoor area on which the sludge residue was piled. Recently, coverage of the area by a greenhouse roof and walls has been added to the design, adding a solar drying effect to the post-treatment process.

Conventionally, sewage sludge is dewatered by mechanical devices, such as decanter centrifuges and screw presses, and subsequently stored until it can be applied on agricultural land as fertiliser (Jensen and Jepsen, 2005). Only a few studies assessing the environmental impacts of sludge treatment technologies include STRB systems (Uggetti et al., 2011; Kirkeby et al., 2013) as data on STRB systems suited for life cycle assessment (LCA) are scarce. Furthermore, the reliability of the results of these studies could be questioned: A considerable part of the inventory data used by Kirkeby et al. (2013) to model the environmental impacts caused by the STRB system strategy were not based on actual data from STRB systems, but on emission data from crop land or compost windrows. Hence, the results presented in Kirkeby et al. (2013) are somewhat unreliable. Furthermore, the LCA method, data and assumptions used in Uggetti et al. (2011) are somewhat intransparent, making it difficult to compare the outcome of that study with other studies. Only in recent years, new life cycle inventory data on STRB systems, including substance flows in STRB systems (Larsen et al., 2017a), gas emissions from the mineralisation process occurring in STRB systems (Larsen et al., 2017b) and fertiliser quality of sludge residue for land application (Gómez-Muñoz et al., 2017), have been generated. Combined, these studies provide the first data set on STRB systems made with the purpose of being suitable for LCA; these datasets were not available at the time the mentioned studies by Uggetti et al. (2011) and Kirkeby et al. (2013) were conducted.

Hence, the aim of the present study was to evaluate the environmental performances of the STRB system strategy using the newest obtained data and according to the international ISO standards for LCA. The performance of the STRB systems strategy was compared to a conventional treatment strategy based on centrifugal dewatering of sludge, subsequent storage and final application on agricultural land. To assure that the comparison of the treatment strategies was up-to-date, new process specific data for the conventional treatment strategy was generated for the purpose. Three sludge treatment scenarios, all of which reflected the management of surplus-activated sludge (SAS) generated at a reference WWTP, were defined and covered treatment, storage, transportation and application of the final sludge product on agricultural land, including the substitution of mineral nitrogen (N), phosphorous (P) and potassium (K) fertiliser, and the treatment of reject water generated during the sludge treatment process.

2. Materials and methods

2.1. The life cycle assessment approach

An LCA can be applied for comparing resource consumption and impacts on the environment of products or services that provide the same function (ILCD, 2010). The LCA in this study complied with ISO 14040 and ISO 14044 standards. An attributional LCA modelling approach was chosen, and the multi-functionality of processes was dealt with by employing system expansion. LCA modelling was done with the mass flow-based LCA software EASETECH, developed by the Technical University of Denmark and as described in Clavreul et al. (2014).

The LCA included three sludge treatment scenarios based on specific case studies of the sludge treatment methods employed at the main reference site, namely a WWTP in Helsinge (Denmark) (56°01′15N; 12°19,49E). This WWTP houses an STRB system, a stockpile area and a mechanical sludge treatment device, namely a decanter centrifuge. Data for the life cycle inventory (LCI) were collected at this site and supplemented by data from three other recent studies (Gómez-Muñoz et al., 2017; Larsen et al., 2017b, 2017a) carried out at Helsinge WWTP and another comparable WWTP, namely Himmark WWTP, (Denmark) (55°2′44″N 9°45′55″E).

Helsinge WWTP receives domestic wastewater and treats it accordingly, corresponding to 25,000 person equivalents (PE) annually. The wastewater is treated by a mechanical-biological wastewater treatment line. A more detailed description of the wastewater treatment line at Helsinge WWTP is found in the Supplementary Information (SI) (section SI-1).

The STRB system at Helsinge WWTP was established in 1996 (Fig. SI-1a and 1b) and is a representative reference for the present-day STRB system technology. Since 1996, it has been well operated, delivering a final sludge residue of high quality. Table SI-1 provides an overview of the operational data and system characteristics for Helsinge STRB.

The stockpile area at Helsinge WWTP was established in 2012–2013 (Fig. SI-1a and 1c) and has a total area of 1675 m^2, with 800 m^2 covered by a greenhouse roof, which enhances evaporation from the sludge residue subjected to treatment. Recently, greenhouse walls on two sides have been added to the design.

2.2. Scope definition

Three sludge treatment scenarios were analysed: 1) mechanical dewatering by a decanter centrifuge, followed by six months of storage, 2) 12 years of treatment in an STRB system and 3) 12 years of treatment in an STRB system followed by four months of post-treatment at a stockpile area. The scenarios were defined as:

Scenario 1: Mechanical treatment (S-CEN).

Sludge is dewatered on a conventional decanter centrifuge and immediately transferred to a container in which the sludge is stored for one week ("on-site storage"). Afterwards, the dewatered sludge is transported 70 km by truck to an external sludge storage facility ("external storage"). Here, the dewatered sludge is laid out in layers of 1–1.5 m in height on the floor in an enclosed storage building. The dewatered sludge is not moved or treated during storage. The storage facility continually receives sludge during the year until autumn, following which it is collected and transferred to a land application site. This procedure implies that at the time of land application, some of the stored sludge has resided at the storage facility for almost one year, while some has only been there for a few days; hence, the average storage time for this study was assumed to be six months. Finally, the dewatered sludge is excavated from the storage facility, transported 200 km by truck to a

land application site and applied by tractor.

Scenario 2: Sludge treatment reed bed system (S-STRB).

Sludge is loaded into an STRB system and undergoes 12 years of treatment (more information on the STRB system technology is provided in SI (section SI-2). The sludge residue (including reeds) is excavated and immediately transported 10 km by truck to a land application site and applied on land by tractor.

Scenario 3: Sludge treatment reed bed system and stockpile area (S-SPA).

Sludge is loaded into an STRB system and undergoes 12 years of treatment (the exact same procedure as in S-STRB). The sludge residue (including reeds) is excavated by an excavator and transported 0.15 km by truck to a stockpile area. The sludge residue is piled and undergoes four months of post-treatment, which is enhanced by solar drying. Finally, the final sludge product is excavated from the stockpile area, transported 10 km by truck to a land application site and applied by tractor.

The system boundaries included all unit processes (Fig. 1) related to sludge treatment and final land application, including the effect of fertiliser substitution, the treatment of reject water and the treatment of SAS produced from the reject water. The temporal scope for the emission inventory and the impact assessment were both defined as 100 years, and the geographical boundary was Denmark. The functional unit (FU) was defined as the treatment and disposal of 1000 kg wet weight (WW) of SAS with characteristics corresponding to the SAS generated at Helsinge WWTP (Table 1). We decided to base the FU on the WW of the SAS as a central purpose of the treatment processes are dewatering, and thereby volume reduction of the sludge. If based on the dry weight of the sludge, this aspect of the treatment processes would not be reflected in the results of the LCA.

Table 1
Quality of the surplus-activated sludge produced at the Helsinge wastewater treatment plant (WWTP).

Wastewater treatment at Helsinge WWTP			
Sludge type		Surplus-activated Sludge (SAS)	
Sludge age (aerobic days)		20–25	
Phosphorous removal		PIX	
Characterisation of surplus-activated sludge (SAS)			
Parameter		Parameter	
Total solid (TS) (% of WW)	0.6790	Cr (% of DW)	0.0023
Volatile solid (VS) (% of DW)	61.483	Mn (% of DW)	0.0747
Total nitrogen (TN) (% of DW)	3.9700	Fe (% of DW)	6.3970
Total carbon (TC) (% of DW)	27.890	Ni (% of DW)	0.0022
NO_3^--N (% of DW)	0.000015153	Cu (% of DW)	0.0314
NH_4^+-N (% of DW)	0.000000001	Zn (% of DW)	0.0573
Mg (% of DW)	0.4234	Cd (% of DW)	0.0001
P (% of DW)	2.2900	Pb (% of DW)	0.0030
Ca (% of DW)	2.8255	K (% of DW)	0.3911

WW: wet weight, DW: dry weight.

2.3. Life cycle inventory (LCI)

2.3.1. Daily operation and transportation

Data and assumptions on daily operations, excavation and transportation included in the various scenarios were based on the present-day situation and procedures at Helsinge WWTP. All scenarios included consumption of electricity due to pumping of sludge and reject water and the consumption of fuel for excavation and transportation. Furthermore, the centrifuging process included in S-CEN requires an additional input of electricity and that the sludge is pre-conditioned by adding polymer coagulant; hence, emissions related to the production of polymer coagulant were included in this scenario. Data on emissions related to the consumption of electricity and fuel in the different scenarios, and the production of polymer coagulants, were taken from the Ecoinvent v 3.3 database and the database included in the EASETECH software. More details on consumption by the three scenarios are to be found in SI (section SI-3).

For S-CEN, it was assumed that the final sludge product was transported 200 km to the land application site, while in S-STRB and S-SPA this distance was only 10 km. Sludge residue treated in a well-operated STRB system commonly meets the threshold values for heavy metals and xenobiotics in biosolids for land application, as required by Danish legislation (Nielsen, 2005; Miljøministeriet, 2017). Furthermore, sludge residue is odourless. Therefore, sludge residue can often be applied on agricultural land in the local area. On the other hand, sludge, which has been mechanically dewatered and subsequently stored, often has a strong odour. Even though the dewatered sludge meets the threshold values required by the

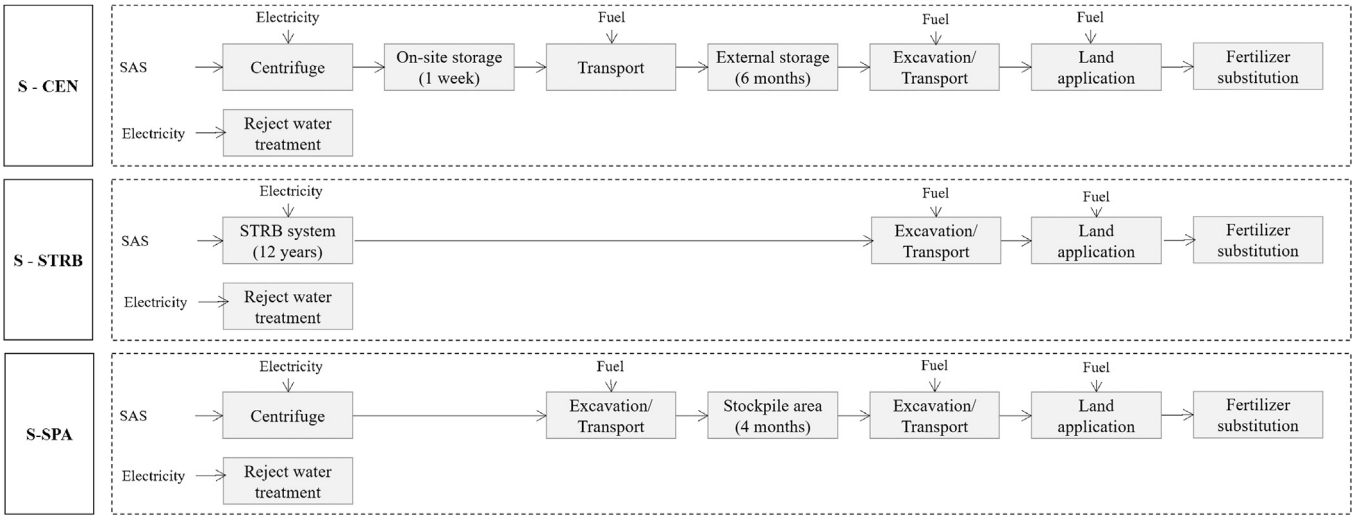

Fig. 1. Unit processes for three sludge treatment scenarios: Scenario S-CEN (dewatering on a centrifuge, one week of on-site storage and 6 months' external storage until land application), Scenario S-STRB (12 years of treatment in an STRB system, excavation in autumn and immediate application on land) and Scenario S-SPA (12 years of treatment in an STRB system, excavation in spring, four months' solar drying at an SPA and, finally, application on land during the following autumn).

legislation, the odour makes it difficult to find a land application site willing to take it (information provided by Grib Vand, the utility managing Helsinge WWTP). Therefore, longer transportation distances are often required, as there are fewer local land application sites available to receive the mechanical dewatered sludge.

2.3.2. Gas emission rates and flow of substances included in S-CEN

Helsinge WWTP houses a centrifuge that is commonly used to treat SAS from other minor WWTPs. To be able to model S-CEN, in which the SAS from Helsinge WWTP is dewatered on the centrifuge, it was arranged that a batch of SAS was dewatered on the centrifuge, instead of being loaded into the STRB, and samples of SAS, dewatered sludge and reject water from the centrifuging process were collected and characterised. These data were used to calculate the amounts of substances allocated to reject water and dewatered sludge, respectively, during the dewatering process. Emission rates for carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) representing on-site and external storage were obtained from flux chamber measurements carried out at the on-site storage facility, a container, at Helsinge WWTP. The dewatered sludge was stored in the container at a height of approximately 1.5 m for 100 days running from October to January. Data on N_2 and ammonia (NH_3) emissions were estimated based on emission data recorded at Helsinge WWTP and data obtained from a study measuring gas emission rates from dewatered sludge piled on an outdoor storage area at a Swedish WWTP (Samuelsson et al., 2018). The Swedish WWTP has a wastewater treatment line that generates sludge comparable to the SAS generated by Helsinge WWTP and was therefore considered an appropriate reference. Gas emission rates, losses of carbon (C) and nitrogen (N) and more information on data sources and calculations can be found in SI (section SI-4).

The evaporation of water during on-site storage was assumed negligible, and during external storage it was estimated by combining our calculated values for losing organic matter and data on the total solids content found in dewatered sludge, before and after 200 days of storage, as published in a publication by the Ministry of Environment and Food of Denmark (Miljø- og Fødevareministeriet, 2000. As this publication reports that no water leached from the sludge residue during storage, and the amount of ash remained unchanged, it was assumed that no P, K or metals had left the system. More information on data sampling procedures, calculations and the shares of substances allocated to different streams in the treatment process can be found in SI (section SI-4). An overview of the flow of substances in the S-CEN scenario based on an input of sludge corresponding to the FU (1000 kg WW SAS) is also provided in SI (section SI-6, Table SI-8).

2.3.3. Gas emission rates and flow of substances included in S-STRB and S-SPA

Losses of C and N by mineralisation, and the related gas emission rates during 12 years of treatment in the STRB system, were modelled using newly generated data presented by Larsen et al. (2017b). In this study, gas emission rates (CO_2, CH_4 and N_2O) from the STRB system in Helsinge, covering all four seasons of the year, were measured by employing static surface flux chambers. Ammonia is produced from NH_4^+ and often constitutes a considerable part of the N loss from sludge and slurries. However, in STRB systems, NH_4^+ is quickly taken up by the reeds or converted into NO_3^- through nitrification, thereby preventing the formation of NH_3. Therefore, it was assumed that the loss of N to NH_3 in STRB systems was negligible. Gas emission rates related to the mineralisation process during four months of solar drying on stockpile area were modelled based on measured emission rates of CO_2, CH_4 and N_2O, also through use of static surface flux chambers. C and N losses, gas emission rates and more information on data sources and calculations are to be found in SI (section SI-5).

The amounts of substances partitioned to reject water, final sludge residue, mineralisation and evaporation during 12 years of treatment in an STRB and the four months of solar drying at a stockpile area, were modelled based on the substance flow analysis presented in Larsen et al. (2017a). The substance flow analysis presented in that study was based on another Danish STRB system, namely Himmark STRB system, albeit it was assumed that the study was an appropriate reference for our LCA, as both systems are run in accordance with the operational guidelines and produce comparable sludge residues of high quality. More information on data sampling procedures, calculations and the shares of substances allocated to different streams in the treatment process can be found in SI (section SI-5). An overview of the flow of substances in the S-STRB and S-SPA scenarios based on an input of sludge corresponding to the FU (1000 kg WW SAS) is also provided in SI (section SI-6, Table SI-8).

2.3.4. Long-term emissions from land application and fertiliser substitution

Emissions related to land application of the final sludge products were modelled using recently obtained emission data for N and C. The emission data representing soil application of SAS treated in an STRB system (S-STRB) originate from a lab-scale soil incubation study presented in Gómez-Muñoz et al. (2017). The fate of C and N over a 100-year modelling period was obtained by using the Daisy soil-plant-atmosphere system model (version 5.21). Gaseous emissions of NH_3 and N_2O, leaching of NO_3^- to groundwater and surface water, N-uptake by crops and C sequestration were estimated. When sludge residues are applied on land, it reduces the need for mineral fertiliser. The environmental savings related to avoiding the production and use of mineral fertilisers were included in the LCA. It was assumed that ammonium nitrate substituted mineral N fertiliser, that single superphosphate substituted P fertiliser and that potassium chloride substituted K fertiliser.

To obtain similar data representing the treatment of SAS in an STRB system combined with post-treatment on a stockpile area (S-SPA) and dewatering by centrifuge (S-CEN), a similar incubation study, following the exact same procedures as described in Gómez-Muñoz et al. (2017), was conducted. More information on the modelling of emissions related to land application and the savings from fertiliser substitution can be found in SI (section SI-7).

2.3.5. Reject water treatment

The reject water generated from the centrifuge in scenario S-CEN or dewatering in the STRB system scenarios S-STRB and S-SPA was returned to the WWTP and treated along with incoming wastewater, thus producing more SAS. All scenarios included one re-run of the reject water, covering its pumping back to the WWTP, the wastewater treatment process (including related emissions to the atmosphere and aquatic environments), the entire sludge treatment processes for the different scenarios and final land application (including fertiliser substitution).

2.4. Life cycle impact assessment (LCIA)

Mid-point impact potentials for 14 normalised impact categories were calculated: Depletion of Fossil Abiotic Resources; Depletion of Reserve-based Abiotic Resources; Climate Change; Marine Eutrophication; Freshwater Eutrophication; Terrestrial Eutrophication; Terrestrial Acidification; Stratospheric Ozone Depletion; Photo Oxidant Formation; Ionising Radiation; Particulate Matter Formation; Human Toxicology – Carcinogenic; Human

Toxicology – Non-carcinogenic and Ecotoxicity. The choice of LCIA methods for the different impact categories was made according to recommendations provided by the International Reference Life Cycle Data System (ILCD) (ILCD, 2010; Hauschild et al., 2013). The normalisation reference was found in (Blok et al., 2013). LCIA methods and normalisation references are shown in SI (section SI-8).

The loadings and savings calculated for each impact category are presented in six sub-processes: 1) daily operation (electricity consumption for pumping sludge and reject water, polymer coagulant consumption), 2) biological gas emissions during treatment and storage (gas emissions related to on-site and external storage of centrifuged sludge or mineralisation processes in STRB system and stockpile area), 3) transportation and excavation (fuel consumption for trucks, excavators and tractors), 4) land application (gaseous emissions and leaching of substances related to land application of the final sludge product), 5) fertiliser substitution (the effect of substituting the production and use of mineral fertiliser) and 6) reject water treatment (RWT) (pumping of reject water back to the WWTP, electricity consumption related to treatment, gaseous emissions and leaching related to treatment, the re-running of the produced SAS through the entire sludge treatment process, including land application and fertiliser substitution).

2.5. Uncertainty analysis

The robustness of the results was analysed on two levels. First, a contribution analysis was performed to identify substances influencing more than 90% of the overall environmental impact (Table 2). Second, a sensitivity analysis (SA) was conducted by increasing and decreasing mineralisation rates and transportation distances for all scenarios. SA-1 tested how increasing or decreasing the C and N mineralisation rates in all scenarios by 10% of its original value affected the outcome of the LCA. SA-2 tested how changing the transport distances affected the outcomes of the LCA. SA-1 and SA-2 were carried out separately, meaning that changes made for the mineralisation of C and N and for transport did not interfere.

3. Results and discussion

3.1. Impact categories

The results of 11 of the 14 impact categories are shown in Fig. 2. The results of the three impact categories not included in Fig. 2 (Stratospheric Ozone Depletion, Photochemical Oxidant Formation and Ionising Radiation) are shown in SI (section S1-9). The impacts of these categories were low compared to the impact categories shown in Fig. 2, and therefore they will not be discussed further.

3.1.1. Climate change, eutrophication and acidification

For the impact category Climate Change, the scenarios provided almost equal net loadings into the environment. The processes resulting in loadings in this impact category were mainly CH_4 and N_2O emissions from treatment in the STRB system (S-STRB and S-SPA), post-treatment at the stockpile area (S-SPA) and storage of dewatered sludge (S-CEN) (Fig. 2 and Table 2). However, in all scenarios emissions of CH_4 and N_2O from the final sludge products after being applied on land also contributed considerable to Climate Change; indeed, for S-CEN almost 50% of the loadings in this impact category was caused by emissions of CH_4 and N_2O related to land application (Fig. 2). Methane and N_2O are strong greenhouse gasses having global warming potentials (GWPs) corresponding to 28 and 265 CO_2 equivalents, respectively, and thus important to consider in relation to Climate Change (IPCC, 2014).

In all scenarios, the treatment processes and the land application processes also emitted CO_2 and N_2. However, as N_2 is not a greenhouse gas and CO_2 originating from biological sources, such as wastewater and sludge, is considered short-cycled C (IPCC, 2007), these emissions are climate-neutral. During the 12-year treatment process in the STRB system in S-STRB and S-SPA, the main share of the mineralised C and N was emitted as CO_2 (93%) and N_2 (94%), while the remaining shares of mineralised C (7%) and N (6%) were emitted as CH_4 and N_2O, respectively. On the other hand, during the six months of storage of centrifuged sludge at the on-site and external storage facilities in S-CEN, only 48% of the C mineralised and 74% of the N mineralised was emitted as climate-neutral CO_2 and N_2, while the remaining shares of C (52%) and N (26%) were emitted as CH_4 and N_2O. The greater production and emission of CO_2 and N_2 from the STRB system (S-STRB and S-SPA) compared to the sludge storage facilities (S-CEN) is assigned to the efficient aeration of the sludge residue in the STRB system: Transportation of air to the sludge residue through rhizomes (hollow out-growths produced by the reeds), movements of stems creating cracks in the sludge residue surface through which air enters and the joint reject water pipe and ventilation system embedded in the filter layer providing air to the lower parts of the sludge residue (Nielsen, 2003). Aeration enhances aerobic microbial activity, leading to the production of CO_2 and N_2. At the sludge storage facilities centrifuged sludge was not moved or turned during the storage period. An earlier study found that anaerobic conditions are prone to develop in dewatered sludge stored in a storage facility without being turned (Nielsen, 2005), leading to production of CH_4 and N_2O.

Table 2
Compounds responsible for >90% of the total impact in 11 impact categories for the three scenarios. The compounds vary among the following six life cycle stages: daily operation, biological gas emissions, transportation/excavation, land application, fertiliser substitution and reject water treatment (RWT).

Impact Category	S-CEN	S-STRB	S-SPA
Climate change	CH_4, N_2O	CH_4, N_2O	CH_4, N_2O
Freshwater eutrophication	PO_4^{3-}, P	PO_4^{3-}, P	PO_4^{3-}, P
Marine eutrophication	NO_3^-	NO_3^-	NO_3^-
Terrestrial acidification	NH_3, SO_x, NO_x	NH_3, SO_x, NO_x	NH_3, SO_x, NO_x
Terrestrial eutrophication	NH_3, NO_x	NH_3, NO_x	NH_3, NO_x
Human toxicity – non-carcinogenic	Zn	Zn	Zn
Ecotoxicity	Zn, Cu	Zn, Cu	Zn, Cu
Human toxicity – carcinogenic	Ni	Ni	Ni
Depletion of fossil abiotic resources	Hard coal, crude oil	Hard coal	Hard coal
Depletion of reserve-based abiotic resources	In, Cd	In, Cd	In, Cd
Particulate matter	NH_3, SO_2	NH_3, SO_2	NH_3, SO_2
Photochemical oxidant formation	NO_x	NO_x	NO_x
	NMVOC	SO_2	SO_2
Stratospheric ozone depletion	CFC-11, CFC-13, HCFC-12	CFC-11	CFC-11

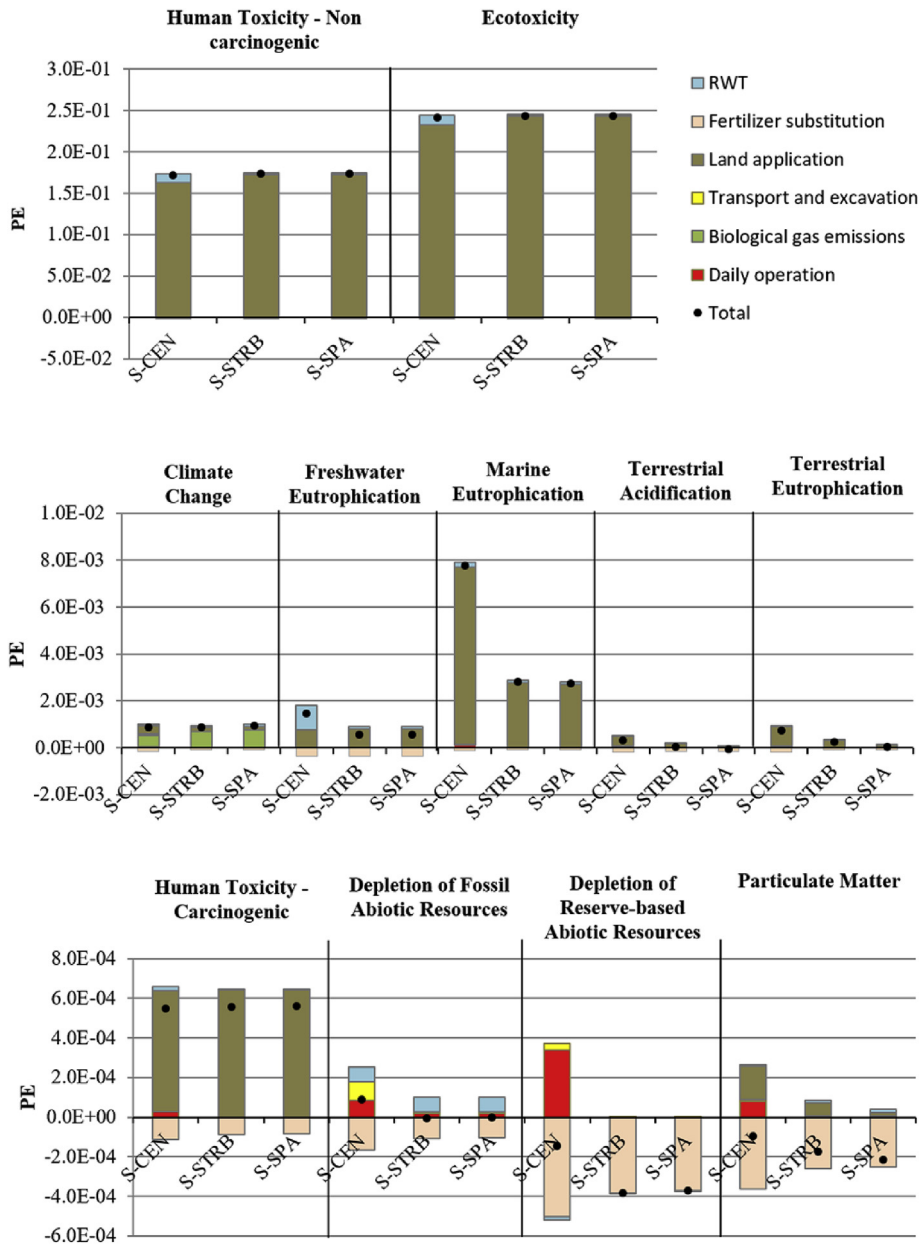

Fig. 2. Life cycle impact assessment of 11 impact categories for three treatment scenarios: 1) dewatering on a centrifuge (S-CEN), 2) 12 years of treatment in STRB (S-STRB) and 3) 12 years of treatment in STRB, followed by four months of post-treatment in a stockpile area covered by a greenhouse roof (S-SPA).

The greater extend of aerobic mineralisation present in the STRB system compared to the storage facilities does not only affect the gas species produced, but also the total amount of C and N mineralised. Aerobic mineralisation is more effective compared to anaerobic mineralisation in terms of the amount of C and N converted into gas species. The amounts of C and N mineralised during treatment in the STRB system were almost twice the amounts mineralised during storage of the centrifuged sludge (Table SI-8), while the emissions to air impacting Climate Change provided by biological gas emissions were almost the same for all three scenarios (Fig. 2). Hence, C and N are more efficiently removed from the sludge treated in S-STRB and S-SPA compared to the sludge treated in S-CEN, despite of the impacts on Climate Change being equal for all three scenarios.

The amount of C and N found in the final sludge product affected the impacts from greenhouse gas emissions related to land application. The slower mineralisation rate during the treatment process in S-CEN meant that more of the C and N was found in the final sludge product, which eventually would be applied on land. Indeed, the greenhouse gas emissions from land application of sludge residue were higher for S-CEN compared to S-STRB and S-SPA (Fig. 2). The share of N emitted as N_2O after soil application was approximately 3% for all three sludge products (SI Table SI-9); however, the N content in the centrifuged, final sludge product was greater compared to the sludge residue from the STRB system, leading to a larger contribution from land application to Climate Change from S-CEN compared to S-STRB and S-SPA. For all scenarios, small environmental savings in Climate Change impact category were obtained by substituting mineral fertiliser.

For Marine Eutrophication, all scenarios showed a net loading, mainly caused by NO_3^- leaching and run-off from land application (Fig. 2 and Table 2). The impact caused by S-CEN was more than

twice the impacts caused by S-STRB and S-SPA. This higher loading in S-CEN was due to the larger N content in the centrifuged sludge, and higher emission factors for NO_3^- leaching and run-off.

For Terrestrial Eutrophication, all scenarios showed a net loading, caused primarily by NH_3 emissions from the land application process (all scenarios) and by NO_x and NH_3 emissions from the combustion of fossil fuels (all scenarios, but especially S-CEN) (Fig. 2 and Table 2). Ammonia emissions were highest in S-CEN as the final sludge produced in this scenario contained more N and had a larger NH_3 emission rate for land application than the final sludge in the other scenarios (S-STRB and S-SPA).

The impact category Terrestrial Acidification was affected primarily by NH_3 emissions from land application and by SO_x,NO_x and NH_3 from the combustion of fossil fuels (Fig. 2 and Table 2). For S-CEN and S-STRB, the overall impacts were small net loadings. For S-SPA, the overall impact was a net saving, as the savings caused by fertiliser substitution exceeded the loadings caused by the other sub-categories.

For Freshwater Eutrophication, net loadings were seen for all scenarios (Fig. 2). For S-STRB and S-SPA, these loadings were caused mainly by phosphate (PO_4^{3-}) leaching from the land application of sludge residues, while for S-CEN the impact potentials caused by PO_4^{3-} leaching from land application and reject water treatment were equal in size (Fig. 2 and Table 2). The concentration of P in the reject water produced by the centrifuge was 10 times the concentration identified in the reject water produced by the STRB. The P leaching factor from the WWTP was relatively high compared to the leaching factor from land application. Therefore, the impact to Freshwater Eutrophication in S-CEN was higher than in S-STRB and S-SPA.

3.1.2. Ecotoxicity and human toxicity (non-carcinogenic)

Among all the impact categories, the most affected were Ecotoxicity and Human Toxicity – Non-carcinogenic (Fig. 2). For both impact categories, all scenarios provided a net loading, caused primarily by the presence of zinc and copper in the final sludge product when applied on land (Table 2). As these impact categories are affected by the same substances, the overall results are the same, except for the magnitude of the values. For both categories, the net loadings were the same for all scenarios. In S-STRB and S-SPA, the loading caused by land application was slightly higher compared to S-CEN, due to the larger amounts of metals transferred to the final sludge product produced in S-STRB and S-SPA (Section SI-6, Table SI-8 in SI). However, metals diverted to the reject water in S-CEN are eventually also land applied, leading to that the contribution to Human Toxicity - Non-carcinogenic and Ecotoxicity were equal for all scenarios.

3.1.3. Human toxicity (carcinogenic), resource depletion and particulate matter

For all scenarios, Human Toxicity – Carcinogenic was affected primarily by the presence of nickel and lead in the final sludge product when applied on land (Fig. 2 and Table 2). Small savings were provided by fertiliser substitution. As for Human Toxicity – Non-carcinogenic and Ecotoxicity, the overall impacts were similar for all scenarios.

The impact of Depletion of Fossil Abiotic Resources (Fig. 2) was higher for S-CEN than for the other two scenarios (S-STRB and S-SPA), due to larger fossil fuel demand related to daily operation, transportation and excavation in this scenario. The main impacts were caused by the consumption of hard coal and crude oil, while the main impacts in S-STRB and S-SPA arose solely from the consumption of hard coal (Table 2). For S-STRB and S-SPA, the overall results were small net savings, as savings from the substitution of mineral fertiliser exceeded loadings. For S-CEN, daily operations included the production and consumption of polymer coagulant required for pre-conditioning the sludge prior to centrifuging. The production of this polymer coagulant caused the consumption of crude oil and the higher environmental loading. Furthermore, the transport distances, earlier addressed in section 2.3.1, included in S-CEN were 70 km from the WWTP to the external storage facility, followed by 200 km to the land application site, compared to 0.150 km from the STRB system to the stockpile area in S-SPA, and 10 km to the land application sites in S-STRB and S-SPA, resulting in a considerably greater demand for fuel in S-CEN.

For the impact category Depletion of Reserve-based Abiotic Resources, all scenarios showed net savings, as the resource consumption avoided from the substitution of mineral fertiliser exceeded the resources needed for sludge management. In S-STRB and S-SPA, positive loadings were negligible compared to savings. For S-CEN, loading was caused mainly by the consumption of lead in relation to the consumption of crude oil. For S-CEN, savings as a result of fertiliser substitution were slightly greater compared to S-STRB and S-SPA, as more mineral fertiliser was assumed to be replaced in this scenario. However, due to the larger loading in S-CEN, this scenario provided a lower net saving compared to the other scenarios.

For Particulate Matter, all scenarios showed net savings, due to the substitution of mineral fertiliser. Positive contributions arose mainly from emissions of NH_3 and sulphur dioxide (SO_2) related to the combustion of fuel, and NH_3 emissions from land application.

3.2. Sensitivity analysis

3.2.1. Mineralisation rates

In SA-1, the mineralisation rates for C and N mineralised were increased and decreased by 10% of their original values in all three treatment scenarios (Fig. 3). Changes in the mineralisation rates during the treatment of sludge in the STRB system, in the stockpile area or while storing mechanical dewatered sludge at the external storage facilities affected the impact category Climate Change, as CH_4 and N_2O emissions were affected (Fig. 3). Furthermore, changing the mineralisation rates affected Marine Eutrophication, as the amount of N found in the final sludge product from the various scenarios depended on the amount of N mineralised earlier in the treatment process. The effects of SA-1 on the remaining impact categories can be found in SI (section SI-10).

For Climate Change, S-CEN was more affected by changes in the mineralisation rates than the other two scenarios (S-STBR and S-SPA) (Fig. 3). The reason for this was that a larger share of the C and N mineralised in S-CEN was emitted as CH_4 and N_2O than in the other two scenarios (Table SI-4 in section SI-4 and Table SI-6 in section SI-5 in SI). When the mineralisation rates for C and N were decreased by 10% of their original values for all scenarios, S-CEN showed a lower Climate Change impact than the other scenarios, while it was higher in the default scenario and when mineralisation rates were increased by 10% (Fig. 3).

Changing the mineralisation rates for C and N had only a small effect on the impact of Marine Eutrophication in the S-CEN (Fig. 3). Due to a much higher mineralisation rate in the STRB system than while storing centrifuged sludge, S-STRB and S-SPA showed changes in Marine Eutrophication. With a 10 % higher mineralisation rate, less N remained in the sludge product, leading to a lower impact on Marine Eutrophication, while with a 10% lower mineralisation rate, more N remained in the sludge product, leading to a higher Marine Eutrophication impact. However, regardless of the mineralisation rate applied, the impact on Marine Eutrophication impact caused by S-CEN was always more than twice as high compared to S-STRB and S-SPA.

The results of SA-1 reflect a trade-off between the impact on

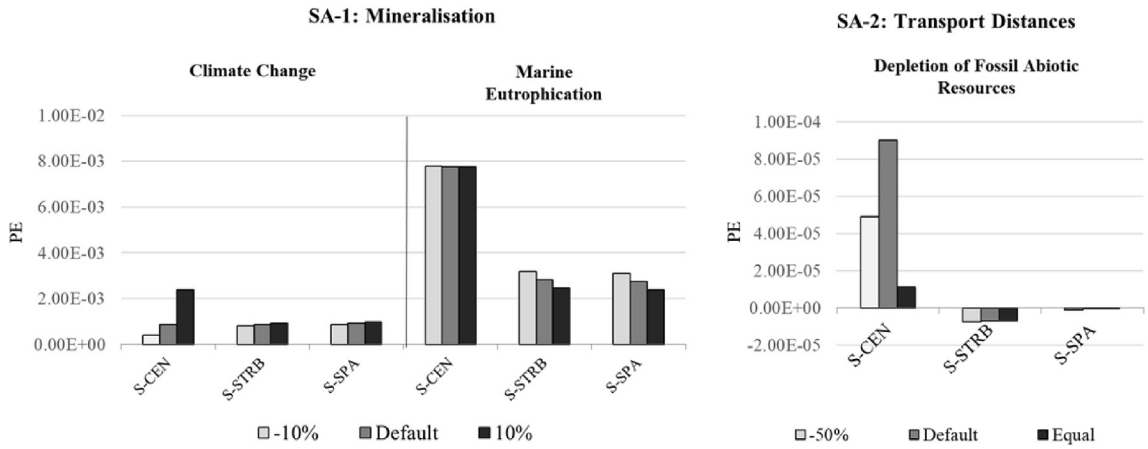

Fig. 3. Results of the sensitivity analysis (SA) testing the robustness of the results in relation to mineralisation rate (SA-1) and transport distances (SA-2) in the treatment scenarios S-CEN, S-STRB and S-SPA. "Default" bars represent total impacts caused by the different scenarios in the LCA modelling. For SA-1, "-10%" and "+10%" represent changes in the impact categories "Climate Change" and "Marine Eutrophication" from the different scenarios, if the amounts of mineralised C and N are decreased or increased by 10%. For SA-2, "-50%" represents the impacts to "Depletion of Fossil Abiotic Resources", if the transport distances in all scenarios are reduced by 50%. "Equal" represents impacts caused if the transport distances in all scenarios are set to 10 km.

Climate Change and on Marine Eutrophication for the mineralisation rates of C and N during treatment or storage: Higher mineralisation rates led to a higher Climate Change impact for S-CEN, but a lower Marine Eutrophication impact for S-STRB and S-SPA, while lower mineralisation rates had the opposite effect.

3.2.2. Transport distances

In SA-2, transport distances in the various scenarios were changed. The total transport distances included in the various scenarios were 270 km for S-CEN, 10 km for S-STRB and 10.15 km for S-SPA. First the transport distances were halved for all the scenarios. Second, the transport distance included in S-CEN was reduced to 10.15 km, to match the transportation distance in the other scenarios. The impact category mainly affected by these changes was Depletion of Fossil Abiotic Resources. Other impact categories affected included Climate Change, Depletion of Reserve-based Abiotic Resources and Particulate Matter. However, the contribution of emissions from transportation in comparison to the emissions from other processes especially biological gas emissions, land application and fertiliser substitution, was minor. The effects of SA-2 on the impact category Depletion of Fossil Abiotic Resources are shown in Fig. 3, while the effects on the remaining impact categories can be found in section SI-10 in SI.

For S-STRB and S-SPA, halving the transport distances did not affect the net impact on Depletion of Fossil Abiotic Resources (Fig. 3), while the net impact for S-CEN was reduced by almost 50%. However, despite this reduction, the impact potential of S-CEN remained higher than for the other scenarios. Reducing the transportation for S-CEN so it equalled the transportation distance in STRB and S-SPA drastically decreased the impact on Depletion of Fossil Abiotic Resources for S-CEN, though it remained greater than in the other two scenarios, due to a high contribution from producing polymer coagulant.

3.3. Discussion

The results of the LCA revealed that in terms of eutrophication of marine and terrestrial environments, the treatment scenarios based on the STRB system strategy (S-STRB and S-SPA) caused lower impacts compared to the conventional strategy using mechanical dewatering on centrifuge (S-CEN). This difference between the treatment strategies are mainly due to that treatment in STRB systems provides a fuller mineralisation of C- and N-containing compounds, without causing a higher emission of greenhouse gasses. None of the strategies are more favourable when considering impacts on climate change; however, the STRB system strategy is favourable in terms of avoiding eutrophication of marine environments in costal zones. Eutrophication of costal zones due to nutrient run-off from agricultural land has during the last decades been a major problem in Denmark, meaning that this difference between the strategies is highly relevant in Denmark and other countries with similar environmental problems.

The LCA study presented in Kirkeby et al., (2013) also found that eutrophication caused by N-containing compounds was higher for sludge treated by a conventional strategy based on mechanical dewatering and subsequent storage compared to sludge treated in an STRB system, though other results, e.g. impacts on climate change, do not comply with the findings of our study. However, due to a lack of data for the STRB system strategy at the time Kirkeby et al. (2013) conducted their study, the results presented in that study are somewhat unreliable, making it difficult to make valid conclusions based a comparing the results to those of our study.

Uggetti et al. (2011), a Spanish study comparing the treatment of sludge in a STRB system with dewatering on centrifuge, concluded, in contradiction with our results, that the impacts caused on Climate Change by emissions of CH_4 from mineralisation of sludge subjected to treatment were negligible compared to the emissions of CO_2 caused by consumption of electricity and fuel. However, Uggetti et al. (2011) did not include emissions of N_2O from the STRB systems, while the results of our study show that emissions of N_2O from mineralisation processes are highly relevant to include for both the STRB system method and the mechanical treatment method. Furthermore, Uggetti et al. (2011) did not include final disposal (land application), as the emissions related to this step were expected to be the same for all scenarios. The results of our LCA suggests that this is not true, but that emissions related to land application, especially those affecting marine eutrophication, are highly relevant when comparing the environmental performances of sludge treatment methods.

Toxic impacts due to heavy metals were found to be the same for all three treatment scenarios. However, the effect of xenobiotics present in the final sludge products were not included in the impact categories addressed in this LCA. The contents of nonylphenol ethoxylates (NPE), di(2-ethylhexyl)phthalate (DEHP), linear

alkylbenzene sulfonates (LAS) and polycyclic aromatic hydrocarbons (PAHs) in sludge products for land application are of concern if the threshold values for these compounds, defined by the Danish Ministry of Environment and Food (Miljøministeriet, 2017), are not met. Hence, the flow of xenobiotics in the treatment scenarios would be a relevant topic for future studies.

Overall, the environmental impacts of S-STRB and S-SPA are almost the same. However, adding post-treatment on a stockpile area to the STRB system strategy has some practical advantages that are not expressed in the results of the LCA. The presence of a stockpile area makes it possible to empty STRB system beds in spring, thereby allowing the reeds in the emptied bed to regrow within a few months, compared to almost one year if emptying happens in autumn. Faster regrowth of the reeds implies that the bed can be reintroduced faster into the loading cycle with a full loading programme, which enhances the treatment capacity of the STRB system. The stockpile area also provides more flexibility in terms of time for emptying and collecting the final sludge product by the recipient.

4. Conclusions

The environmental impacts caused by the sludge treatment scenarios based on the sludge treatment reed bed (STBR) system strategy (S-STRB and S-SPA) were comparable or lower than the scenario representing a conventional sludge treatment strategy including mechanical dewatering on a centrifuge and subsequent storage (S-CEN). Carbon (C) and nitrogen (N) was more efficiently removed by the treatment processes included in S-STRB and S-SPA, resulting in a lower content of C and N in the final sludge product compared to S-CEN. The lower content of C and N in the final sludge product produced by S-STRB and S-SPA resulted in considerable lower impacts in terms of marine eutrophication compared to S-CEN. The impacts on climate change caused mainly by gas emissions from the treatment process were equal for all scenarios. The impact on resource depletion was higher for mechanical dewatering due to a larger fossil fuel demand related to daily operation and longer transportation distances in this scenario. A sensitivity analysis revealed that the performances of S-STRB and S-SPA were more robust to changes in the amounts of C and N mineralised during the treatment process and changes in transport distances compared to S-CEN. In terms of human toxicity and ecotoxicity, the impacts for all three treatment scenarios were comparable. According to the results of the LCA, there were no considerable differences in the performances of S-STRB and S-SPA. However, adding a stockpile area to the STRB system strategy has some practical advantages, which should be considered.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.jclepro.2018.02.193.

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